

### **REMARKS / ARGUMENTS**

The summary page of the Office Action dated December 28, 2007, indicates that the Action is nonfinal, but the body of the Office Action states that it is final. In a brief telephone interview initiated by applicants' undersigned attorney on June 23, 2008, for the sole purpose of clarifying whether the Action is final, the Examiner confirmed that the Action is indeed final. This response and accompanying submissions have been prepared accordingly.

#### ***Claim Rejections 35 USC § 103***

First of all, before addressing the Examiner's remarks in detail, it is reiterated that the crux of the present invention is to produce a reaction between molten aluminum and an additive to form a dense phase in the pores that prevents leaching of  $TiB_2$  from the carbonaceous component. If the amount of  $TiB_2$  initially introduced as such into the carbonaceous component is too low, e.g. if the process of Bergeron et al. is followed using 10% by weight or less of  $TiB_2$ , the surface of the component will not be wettable by molten aluminum and the molten aluminum will not enter the pores and react with the additive to produce the desired dense phase. There is no teaching of this in the prior art. In fact, none of the references even recognizes the specific problem of leaching of metal boride, much less that the problem could be solved by reacting molten aluminum with an additive, and that this reaction would result from the combination of the presence of the additive and of sufficient initial  $TiB_2$  to provide initial wettability. It is therefore believed that the present invention is unobvious from the references taken alone or in any combination.

The specific remarks made by the Examiner are now addressed using the same marginal numerals.

**3&4.** The Examiner referred to page 3 of Applicant's Remarks in the previous response of September 27, 2007 and commented that the arguments with regard to the "admitted prior art" are not convincing. However, it seems that the Examiner may have misinterpreted what was being said. The comment in question is believed to be the following:

The "admitted prior art," as applied in the final rejection, merely "shows that a process for making cathodes for an aluminum reduction cell by forming a composite of a carbon-containing component and a metal boride such as titanium diboride is known"; hence, the

“admitted prior art” does not teach the combination of sufficient  $\text{TiB}_2$  to make the surface wettable by aluminum with any additive. (Underlining added)

The point being made here is that the “admitted prior art” does not teach the need for a combination of sufficient  $\text{TiB}_2$  to make the surface wettable with an additive of any kind. All that the “admitted prior art” discloses is that it was already known to use metal borides, such as  $\text{TiB}_2$ , in carbon-containing components to protect the cathode against erosion and oxidation and to make the surface wettable. There is no disclosure or suggestion of the need for any additive to accompany the  $\text{TiB}_2$ .

The Examiner stated: Thus, the “admitted prior art” does recognize the inclusion of sufficient  $\text{TiB}_2$  to make the surface wettable. This may be so (although there is no reference to the use of at least a minimum amount). However, there is no suggestion in the “admitted prior art” of a minimum or maximum amount of  $\text{TiB}_2$  for wettability in combination with precursors of  $\text{TiB}_2$  or additives.

As stated previously, Bergeron et al. describes a combination of an additive mixture (“precursor oxide mixture”) with carbonaceous material that includes at most only a small amount (3-10%) of  $\text{TiB}_2$  powder to increase the kinetic of  $\text{TiB}_2$  formation during cell operation. This amount of  $\text{TiB}_2$  is not enough to make the surface of the carbonaceous material wettable. To demonstrate this fact, Applicant is attaching a second Declaration from Dr. Brisson. The Declaration describes tests that were carried out to demonstrate, *inter alia*, that there is a minimum amount of  $\text{TiB}_2$  as such required to make the surface wettable. The minimum amount is somewhere between 15 and 20% by weight (15% was insufficient, whereas 20% was sufficient). This is consistent with the teachings of Bergeron et al. (30% provides wettability) and Townsend (20% is about the minimum). The minimum level is thus higher than the maximum amount of  $\text{TiB}_2$  (10% by weight) suggested by Bergeron et al.

The Examiner acknowledges that, while this may be correct (which the enclosed Declaration confirms), one of ordinary skill in the art would be able to determine the amount of  $\text{TiB}_2$  necessary to achieve wettability. Clearly this is so because Bergeron et al. and Townsend both disclose amounts suitable for wettability. However, what the person of ordinary skill in the art would not be able to derive from Bergeron et al. (or Townsend) is that, if you wish to obtain the benefits of the present invention (reduction of leaching of borides), one must have (a) sufficient  $\text{TiB}_2$  (as such) present in the mixture from the start (20% or more), and (b) provide an

additive to react with the molten aluminum to seal the pores. As already noted above, if the process of Bergeron et al. is followed, the amount of  $\text{TiB}_2$  would be insufficient to make the surface wettable and, therefore, there could be no reaction between aluminum and the additive in the pores to produce the desired sealing effect.

5. The Examiner went on to say that Bergeron et al. provides substantially all the titanium diboride in the form of precursors which are reacted *in situ* so as to maximize cost savings, whereas the "admitted prior art" shows that all  $\text{TiB}_2$  may be provided as  $\text{TiB}_2$  itself rather than as precursors. The Examiner stated:

Choice of an amount of  $\text{TiB}_2$  between these two recognized values would have been obvious to one of ordinary skill in the art. By using an amount of additives less than the maximum, some amount of savings would still have been achieved.

Applicant strongly disagrees with this statement. As stated in MPEP, Section 2141.02, paragraph VI, a prior art reference must be considered in its entirety, i.e., as a whole, including portions that would lead away from the claimed invention. Bergeron et al. states on page 3, at lines 20 to 24 that:

Although titanium boride is preferred from the point of view of superior performance, it has the considerable disadvantage that it is very expensive. At present, for example, the cost of  $\text{TiB}_2$  is approximately CDN\$66,180 per metric ton (approximately CDN\$60,000 per ton).

On page 5, at lines 18 to 20, it is stated that:

The cost of such metal oxides and boron oxides (in boric acid form) is about one thirtieth of the cost of equal amounts of metal borides, e.g.  $\text{TiB}_2$ .

On page 7, lines 18 to 28, it is stated that:

In contrast to the CDN\$66,180 per metric ton (CDN\$60,000 per ton) mentioned above as the cost of  $\text{TiB}_2$ , an equivalent amount of a precursor oxide mixture currently costs in the region of CDN\$2,206 per metric ton (CDN\$2,000 per ton). The cost savings achievable by the present invention is clearly significant. When the invention is used to form wettable electrodes for drained cathode cells, the economic advantage obtained during the

preparation of aluminum, compared to the use of blocks prepared from  $\text{TiB}_2$ , has proven to be about CND\$66 per metric ton of metal (CND\$60 per ton of metal).

The very clear teaching of Bergeron et al. is therefore that precursors of  $\text{TiB}_2$  are very much less expensive than  $\text{TiB}_2$  itself, so that there is every motivation to use as little  $\text{TiB}_2$  as possible.

Nevertheless, it is stated on page 13, lines 7 to 11 that:

A certain amount of  $\text{TiB}_2$  powder (e.g. 3% - 10%) with a particle size smaller than  $15\mu\text{m}$  in the paste during mixing, could be added to the paste during mixing to increase the kinetic of  $\text{TiB}_2$  formation during cell operation.

In the case of EXAMPLES 1 and 2 of Bergeron et al., there are statements that "To this paste was added about 3-10% by weight of  $\text{TiB}_2$  powder" to each of three formulations. In the case of EXAMPLE 3, there was mention of the use of "a small quantity of  $\text{TiB}_2$  in the sample before forming and baking." Claim 2 of Bergeron et al. on page 19 allows for an amount of 3 to 10% by weight of metal boride.

Therefore, throughout Bergeron et al. there is an emphasis on replacing all of the  $\text{TiB}_2$  with oxide precursors for the sake of cost saving except for "a small quantity" (i.e. 3 to 10% by weight) of the metal boride required to improve the kinetics of boride formation during cell operation. A skilled person after reading Bergeron et al. would not think of using more than the suggested 10% by weight of  $\text{TiB}_2$  because the entire emphasis of the reference is to use none, or as little as necessary to achieve the stated objective (which requires no more than 10%). Thus, a person skilled in the art, upon reading Bergeron et al. as a whole would see no reason to use more than 10% by weight of  $\text{TiB}_2$ . There would be no logic in doing so. If the motive is cost saving, clearly the more that can be achieved, the better. The Examiner suggests that any amount of replacement of boride by precursors would achieve some cost saving, but there is no reason why a person skilled in the art would opt for a minor cost saving rather than a major one when the latter could be had with no greater difficulty than the former.

It is also to be noted that Bergeron et al. teaches (see page 3, lines 20-26) that the boride formed *in situ* from the precursors themselves (ultimately) provides acceptable wettability. Hence, the desire for wettability would not provide motivation to increase the initial amount (and

cost) of  $\text{TiB}_2$  suggested by Bergeron et al. Also, in view of this teaching, there would be no reason to limit the substitution of cost-saving Bergeron et al. precursors for  $\text{TiB}_2$  itself (e.g. in the materials of Townsend and “the admitted prior art”).

Further, the claims of the present application are limited to a maximum of 25% by weight of the additive. There is nothing in Bergeron et al. to suggest such an upper limit on the use of boride precursors. On page 13 of Bergeron et al., the ratio of precursor oxide mixture to carbonaceous material is “normally in the range of 40:60 parts by weight, more preferably 50:50 parts by weight.” This is clearly above the amount specified in the claims of the present application.

It should also be noted that the present invention provides unexpected results in the form of reduction of leaching of the boride from the carbonaceous material. This is proved by the Examples of the present application. There is no mention of this in any one of the “admitted prior art”, Bergeron et al. or Townsend. In the absence of such knowledge of unexpected results, there would be no reason for a person skilled in the art to select the range of additive required by the claims of the present application.

It is also pointed out that the enclosed Declaration shows that a further unexpected result is the improved erosion resistance obtained by the additions of the additive over and above that produced by the additions of  $\text{TiB}_2$  (see Figure 5 of the Declaration). Again, this is not to be expected from the teaching of the prior art.

As stated earlier in the prosecution of this application, even assuming *arguendo* that the method of claim 1 is *prima facie* obvious (which is denied), it is well settled that *prima facie* obviousness of a novel combination may be overcome if the combination achieves unexpected beneficial results. It has been demonstrated above that this is the case.

It is therefore believed that the claimed invention is not obvious from the references cited by the Examiner and reconsideration is requested.

6. The Examiner also commented on the Declaration submitted previously stating that it is not commensurate in scope with the claimed subject matter. The Examiner pointed out, *inter alia*, that the Declaration exemplified only  $\text{TiO}_2/\text{B}_2\text{O}_3$  as the additive whereas the claims recite an additive consisting of two intimately mixed compounds without specifying what the compounds are. The Examiner therefore objected that Claim 1 is of greater scope than Applicant's evidence.

In view of this, the claims have been limited to mixtures of  $\text{TiO}_2/\text{B}_2\text{O}_3$  as the additive. There is no intention on the part of the Applicant to abandon the broader scope of the invention as originally claimed. This amendment is being made at this time merely to make the claimed scope more commensurate with the evidence presented. Broader claims may be pursued in the future by way of a continuation or divisional application.

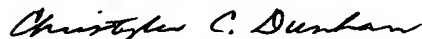
In view of the above arguments and amendments, favorable reconsideration of this application is requested.

Respectfully,



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I hereby certify that this paper is being deposited this date with the U.S. Postal Service as first class mail addressed to Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.



Christopher C. Dunham  
Reg. No. 22,031 Date JUNE 26, 2008



*Docket No. 71624 CCD*

**IN THE UNITED STATES  
PATENT AND TRADEMARK OFFICE**

**APPLICATION NO** : 10/723,966  
**APPLICANT** : DIONNE, Martin; et al.  
**FILED** : November 26, 2003  
**TITLE** : STABILIZERS FOR TITANIUM DIBORIDE-  
CONTAINING CATHODE STRUCTURES  
**ART UNIT** : 1742  
**EXAMINER** : LEADER, William T.

**DECLARATION UNDER 37 C.F.R. § 1.132**

I, Dr. Pierre-Yves BRISSON, declare that:

1. I have been a full-time employee of Alcan International Limited, the assignee of the present application, since January 1, 2006, and I was employed under contract to the same company from June 2002 until completion of my Ph.D. My current position with the company is Research Scientist in the Strategic Research Group working on materials for wettable cathodes.

2. I have a Ph.D. in Chemical Engineering from the Université de Sherbrooke, Québec, Canada. The subject of my thesis was the study of sodium and bath penetration mechanisms within the carbon cathode of the aluminum electrolysis cell. Consequently, the subject of my Ph.D. is relevant to studies on deterioration of carbon materials (including those containing titanium diboride) in electrolysis cells. I also hold the degree of M.Sc.A., Metallurgical Engineering, from École Polytechnique de Montréal, 2002. The subject of my master's thesis was the understanding of the relationship between the microstructure of refractory materials and their mechanical behaviour at high temperatures (1200 to 1400°C). I also hold the degree of B.Eng., Materials Sciences and Engineering, from École Polytechnique de Montréal, 2002.

3. I was asked to devise and perform a number of experiments as described below. This required me to produce cathode blocks in the laboratory and to evaluate them in terms of their wettability with molten aluminum and erosion resistance under electrolysis. The objective of this work was to provide a clear example showing the impact of using a stoichiometrical mixture of powders of  $\text{TiO}_2$  and  $\text{B}_2\text{O}_3$  of 75 to 175 microns (referred to below as "Mixture M" for simplicity) in wettable cathode blocks. More specifically, the work aimed at first finding the minimum amount of  $\text{TiB}_2$  to

reach complete wettability, and then to evaluate the impact of adding a small amount and a larger amount of Mixture M in these blocks.

### 3.1. Experimental Set-Up

First, a very broad set of block compositions was prepared all at once to cover all possibilities (see Table 1 below). The goal of this experimental work was to first establish the minimum amount of  $TiB_2$  required to reach wettability. Since it is known that 30% of  $TiB_2$  leads to wettability (see Canadian patent 2350814 to Bergeron, page 10, lines 7 to 9) blocks with 15, 20 and 25% of  $TiB_2$  were prepared. After this, to measure the effect of additions of Mixture M, blocks were produced with 15 to 30 wt.% of  $TiB_2$  and 10 or 25% of Mixture M.

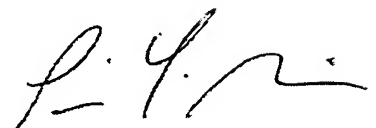
It is to be mentioned that the amount of pitch was not optimized for these blocks. The amounts used here were believed to be close to optimum based on past experience. The carbon particle size distribution used in these blocks is presented in Table 2 below. All blocks were baked for 5 hours at 1100°C and cores were drilled for testing under electrolysis and physical characterization. The measured properties were electrical resistivity, compressive strength and bulk density.

Table 1  
List of the blocks prepared

#	$TiB_2$ (%wt)	Mixture M (%wt)	Pitch (%wt)	Anthracite %	Green Density (g/cm <sup>3</sup> )
570	15	---	14.6	Balance	1.68
567	20	---	15.0	Balance	1.75
568	25	---	15.0	Balance	1.87
578	15	10	14.0	Balance	1.84
577	15	25	14.0	Balance	1.93
569	20	10.0	15.5	Balance	1.89
572	20	25	15.0	Balance	1.99
576	20	25	14.0	Balance	2.00
571	25	10	14.6	Balance	1.96
573	25	25	15.0	Balance	2.05
575	30	10	13.0	Balance	2.02
574	30	25	14.0	Balance	2.14

Table 2  
Carbon particle size distribution

Size (mesh)	Amount (%)	
	FREQ.	CUM.
-3+4M	17	100
-6+8M	41	83
-8+14M	22	42
-14+28M	9	20
-28+48M	2	10
-48+100M	2	8
-100+200M	2	6
-200	4	4





### 3.2. Electrolysis tests

For the testing under electrolysis, two different types of tests were done. The first one, the so-called "monopod", is depicted in Figure 1 below (left hand side). This test was mainly used to evaluate the block's wettability and behaviour under electrolysis. The cathode specimens were drilled out of the block and put in the electrolyte. The graphite crucible acted as the anode. The tests conditions are presented in Table 3 below. At the end of the test, the samples were cut into slices and analyzed for erosion and wettability using image analysis tools (erosion was measured by comparing the cross sectional area of the samples before and after electrolysis, while wettability was measured as the percentage of coverage of the surface by aluminium). The second test (the so-called "rotating cathode" test) was used to estimate the erosion. This test was described in details in my previous Declaration of September 20, 2007 submitted in these proceedings. A photograph of the set-up is presented in Figure 1 below (center and right hand side) and the test parameters are presented in Table 3 below. These trials were carried out in parallel to obtain as much information as possible from the materials in the shortest possible amount of time.



**Figure 1**  
**The « Monopod » and « Rotating » set-ups**

**Table 3**  
**Electrolysis tests parameters**

Parameter	Unit	Rotating	Monopod
Temperature	(°C)	970	970
Ratio	---	1,1	1,1
CaF <sub>2</sub>	(%)	5	5
Alimentation	(g/h)	40	~10
Rotation speed	(RPM)	30	0
Current	(A)	65	90
Length	(h)	100	100
Immersion	(cm)	6	5
ACD	(cm)	2.5	Crucible

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#### 4. Results and discussion

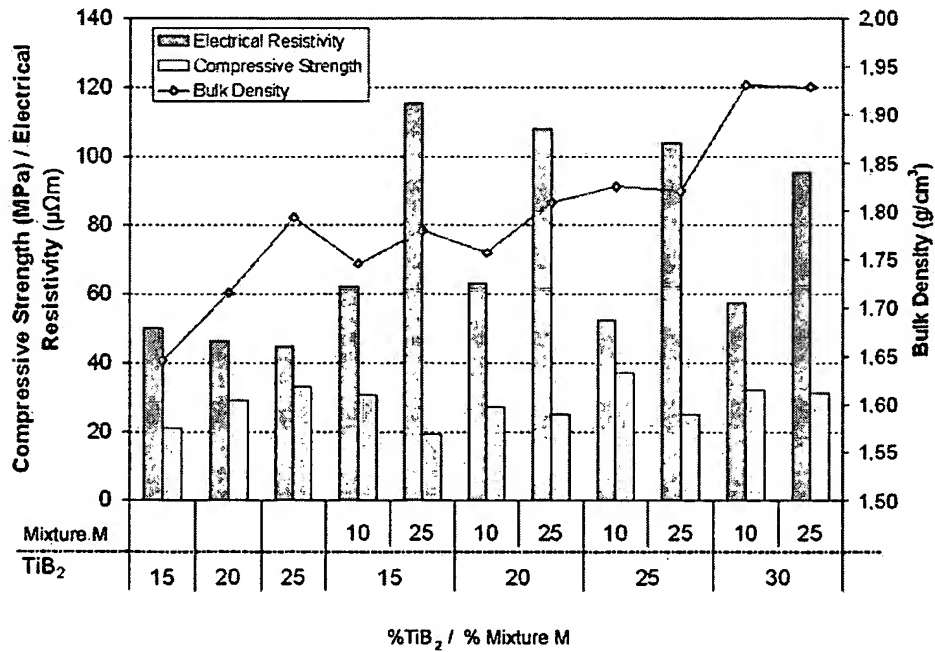
The results of the tests are discussed below.

##### 4.1. Materials properties

The results obtained are presented in Table 5 and Figure 2 below.

**Table 5**  
**Materials Properties**

#	TiB <sub>2</sub> (%wt)	Mixture M (%wt)	Bulk Density (g/cm <sup>3</sup> )		Electrical Resistivity (μΩm)		Compressive Strength (MPa)	
			Average	S.D.	Average	S.D.	Average	S.D.
570	15	---	1.65	0.006	50.5	0.6	21.3	0.96
567	20	---	1.72	0.003	46.7	0.5	29.3	1.93
568	25	---	1.79	0.002	45.1	0.5	33.2	1.73
578	15	10	1.75	0.003	62.1	0.2	30.9	0.68
577		25	1.78	0.011	115.3	8.4	19.3	5.59
569	20	10	1.76	0.009	63.0	0.9	27.4	0.51
572		25	1.81	0.015	108.1	7.0	25.3	1.29
571	25	10	1.83	0.003	52.5	3.8	37.4	5.94
573		25	1.82	0.009	103.8	7.8	25.3	3.76
575	30	10	1.93	0.008	57.5	1.9	32.3	1.33
574		25	1.93	0.012	95.4	9.0	31.4	6.33



**Figure 2**  
**Properties of the Materials as a function of composition**

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## 4.2. Wettability and Erosion Measurements

### 4.2.1 Evaluation of the minimum $\text{TiB}_2$ requirement for wetting

Figure 3 below presents a typical view of the samples with 15 to 25%  $\text{TiB}_2$  after 100 hours of electrolysis. The corresponding measured wetting indexes are indicated in Table 6 below along with the erosion measurements. As one can see, there is little to no difference between the block with 20% of  $\text{TiB}_2$  and the one with 25%, in terms of wetting and erosion, while the block with 15%  $\text{TiB}_2$  is clearly deficient in wetting and exhibits higher erosion. This being said, all samples exhibit somewhat high erosions, as the very rough surface of the samples suggests. Because the decision to continue with one material was to be based on the minimum wettability, the next electrolysis tests were performed on the materials having 20% of  $\text{TiB}_2$  and 10 or 25% of Mixture M.



Figure 3

Cross sections of the materials with 15, 20 and 25% of  $\text{TiB}_2$  after 100 hours of electrolysis

Table 6

Wettability and erosion of the materials without Mixture M after 100 hours of electrolysis

Sample #	% $\text{TiB}_2$	Wettability (%)		Erosion (mm)	
		Average	S.D.	Average	S.D.
570	15	68.6	8.96	3.9	0.09
567	20	96.4	4.28	3.1	0.57
568	25	92.6	3.91	2.8	0.17

### 4.2.2 Impact of adding Mixture M on wettability and erosion

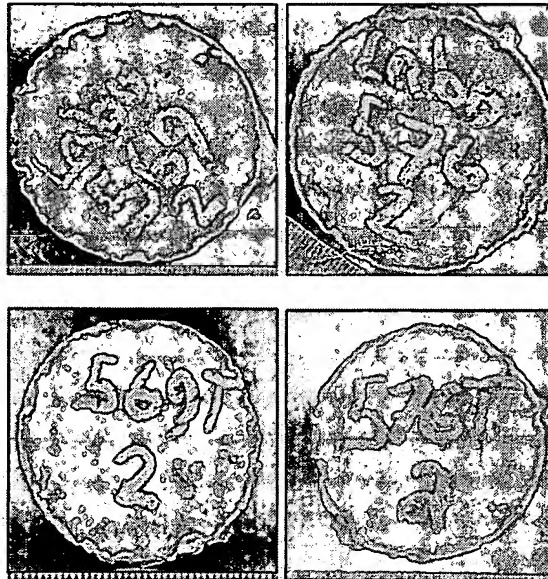
Table 5 above shows that close to 100% wettability is achieved with 20% wt and 25% of  $\text{TiB}_2$  but that erosion remains somewhat high. Table 6 below presents the impact of adding Mixture M in these materials on wettability and erosion while Figure 4 below presents a typical cross-section of the samples after electrolysis (monopod and rotating). The results from Table 7 below are also presented in graphical form in Figure 5 below.

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**Table 7**  
**Wettability and erosion of the materials with Mixture M**  
**after 100 hours of electrolysis**

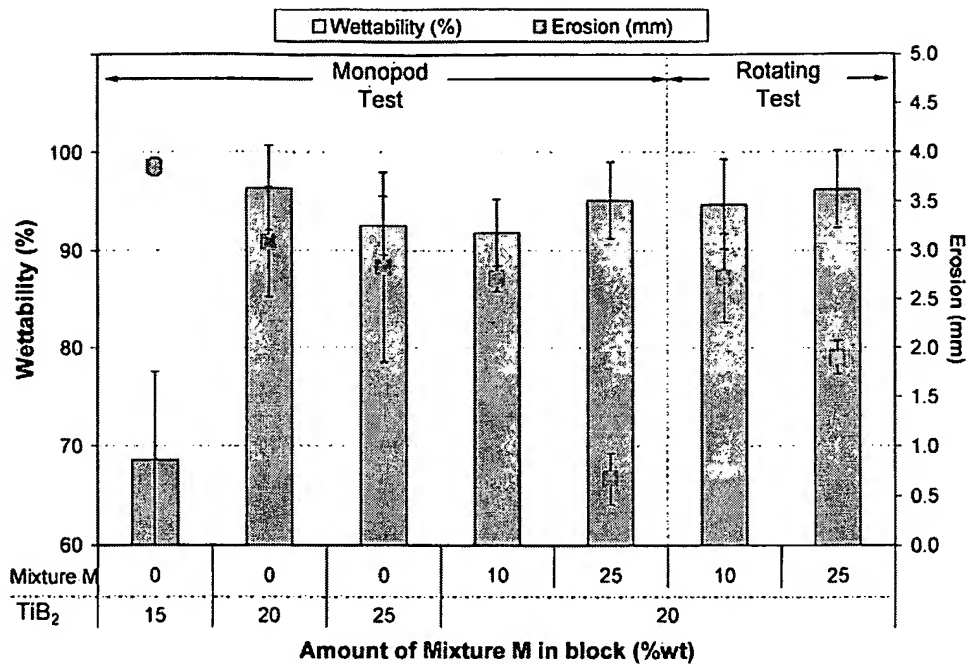
Sample #	TiB <sub>2</sub> (%wt)	Mixture M (%wt)	Wettability (%)		Erosion (mm)	
			Average	S.D.	Average	S.D.
570	15	---	69	9.0	3.9	0.09
567	20	---	96	4.3	3.1	0.57
568	25	---	93	3.9	2.8	0.17
569	20	10	92	3.0	2.7	0.98
576	20	25	95	3.4	0.7	0.13
569T*	20	10	95	3.9	2.7	0.26
576T*	20	25	96	4.6	1.9	0.46

\*The "T" stands for the "Rotating Test"



**Figure 4**  
**Cross sections of the materials with Mixture M**  
**100h of electrolysis with and without rotation**

*P. J. R.*



**Figure 5**  
**Measurements of the wettability and erosion of the materials**  
**after 100h of electrolysis**

It appears that wettability does not vary much with the addition of Mixture M. In fact, all the data fell within the measures dispersion. One can thus conclude that at this level of wettability Mixture M has a negligible effect. This being said, the wettability measured on the rotating samples is high compared with other tests realised lately on samples without Mixture M – which is here a strong argument in favour of the desirable impact of Mixture M on wettability.

The values obtained from the erosion measurements clearly suggest a positive impact of Mixture M. Although these measurements usually lead to somewhat high dispersion in the measured values, both methods indicate a lower erosion of the block with 10% of Mixture M and even distinctly lower erosion with 25% of Mixture M. Thus even if caution is recommended in the analysis of these data, the confidence in these results is high. Note that the higher erosion measured in the rotating test, as compared to the monopod test, is related to the more aggressive conditions of the former.

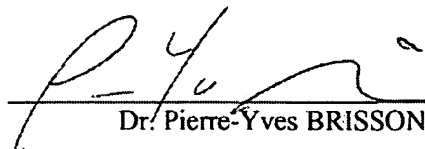
## 5. Conclusions

In order to evaluate the impact of Mixture M on the behaviour of the wettable blocks under electrolysis, carbon blocks were produced in the laboratory with selected amounts of TiB<sub>2</sub> and Mixture M. The first goal was to identify the minimum amount of TiB<sub>2</sub> required to reach a near to complete wetting. The results showed that no differences were noticeable in terms of wetting between 20 and 25% of TiB<sub>2</sub>. When Mixture M was added to the blocks already containing 20% of TiB<sub>2</sub>, wetting was not improved significantly (since it was already over 95% of coverage). However, two different test set-ups lead to the conclusion that as little as 10% of Mixture M is

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sufficient to improve erosion resistance and that better erosion resistance was obtained when 25% of Mixture M was added instead of 10%.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

  
Dr. Pierre-Yves BRISSON

Date: 18-06-08